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Experimental investigation of picosecond dynamics following interactions between laser accelerated protons and water

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We report direct experimental measurements with picosecond time resolution of how high energy protons interact with water at extreme dose levels (kGy), delivered in a single pulse with the duration of less than 80 ps. The unique synchronisation possibilities of laser accelerated protons with an optical probe pulse were utilized to investigate the energy deposition of fast protons in water on a time scale down to only a few picoseconds. This was measured using absorbance changes in the water, induced by a population of solvated electrons created in the tracks of the high energy protons. Our results indicate that for sufficiently high doses delivered in short pulses, intertrack effects will affect the yield of solvated electrons. The experimental scheme allows for investigation of the ultrafast mechanisms occurring in proton water radiolysis, an area of physics especially important due to its relevance in biology and for proton therapy. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4977846>]

The field of laser based ion acceleration has made rapid progress in the past decade.^{1,2} As control over ion beam parameters increases, it becomes possible to find and develop applications utilizing their unique properties. Ion pulses from laser particle accelerators are created on the time scale of a picosecond,³ and although they commonly have a broad energy spectrum, close to their source they are significantly shorter⁴ than the ion beams from conventional accelerators. Furthermore, it is possible to synchronise multiple laser pulses, provided they come from the same source, with time jitter that is negligible on the picosecond time scale, and thus enabling the possibility to use one laser pulse to drive the acceleration process and one as a probe, in a pump/probe-configuration.⁴ Until now, studies have been conducted probing or imaging the actual acceleration process with shadowgraphy or using the accelerated protons to probe laser generated dense plasmas.⁵ In this paper, we present results where the short pulse duration, in combination with the exceptional synchronisation properties, is utilized to investigate how energetic protons interact with water with a time resolution of only a few picoseconds.

The experimental research of proton radiolysis in water has so far been limited in time resolution by available proton pulse durations and by electronic synchronisation between the proton pulse and a separate probe. This has made it impossible to make direct experimental studies of processes that are faster than a few hundred picoseconds. Another option is to use indirect scavenging techniques, where a chemical scavenger is employed to measure the yields of radiolytic species in the tracks of high energy protons.⁶ By varying the concentration of the scavenger, the yields can be accurately determined at different times using the rate constant for the reaction

between the scavenger and the radicals. For picosecond time scales, however, this method becomes less reliable due to the very high concentration of scavengers needed. Traditionally, such experiments were performed in combination with Monte Carlo simulations^{7,8} to study the early effects of the high energy protons on water.

As high energy protons propagate into water, the absorbance of the water changes for certain wavelengths. This process occurs when the protons deposit energy through ionising the water molecules and thereby releasing electrons. In the water, these secondary electrons will first become thermalized and then subsequently solvated/hydrated and, among other things, have the property of absorbing light in the visible and near-infrared region.⁹ Even though the existence of solvated electrons has been known for decades, their exact properties and behaviour are not fully understood and are to some extent still debated.^{10–12} From a biological perspective, the study of how high energy protons interact with water is highly relevant since as much as 66% of radiation deposited into a cell is initially absorbed by water molecules. The radicals and solvated electrons formed in this interaction can then react with and damage the DNA in the cell.¹³

In the proton pulse radiolysis experiment we present here, the protons were accelerated from thin aluminium foils in the target normal sheath acceleration (TNSA) regime.^{14,15} Here, the leading edge of an ultrashort high intensity laser pulse fully ionises the front surface of a target foil and turns it into a plasma. The ponderomotive force of the laser pulse pushes electrons through the plasma and sets up a quasi-static electric field on the back of the foil. Contaminations, such as hydrocarbons and water, which can be found here become ionised and preferentially the protons (H^+) are accelerated in the electric field to MeV-energies. This gives a characteristically broad, exponentially decaying, and in our

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case, non-relativistic proton energy spectrum, meaning that the protons within a pulse travel with different velocities. This proton pulse is also divergent. Due to these factors, it is important to study the proton-water interaction as close to the proton source as possible, to fully take advantage of the short proton pulses and also to reach the highest dose levels.

The experiments were performed at the TARANIS laser at Queen's University, Belfast, UK. The laser delivered 10 J in 600 fs long pulses with a central wavelength of 1053 nm and a repetition rate of approximately 1 pulse per 10 min. The laser pulses were focused with an $f/2$ off-axis parabolic mirror onto a 12 μm thick aluminium foil at 30° angle of incidence. Protons were accelerated to energies up to ~ 10 MeV, which was measured with stacks of radiochromic film,¹⁶ that change color when absorbing radiation. 1 cm behind the target a water-cell was placed (see Fig. 1). The optical absorbance of the water in this cell was probed, perpendicular to the proton propagation direction, with a 1 ns long chirped laser pulse, originating from the same source as the main pulse. The chirp in the probe pulse was introduced in a double pass grating stretcher and was tuned in a dedicated grating compressor. In the beam path of the probe pulse, two mirrors were placed on a translation stage so that the relative delay between the proton pulse and the probe pulse could be varied over a few ns. The interaction area was imaged by a lens with 8 times magnification onto the entrance slit of a 1 m imaging Czerny-Turner spectrometer with a $10 \times 10 \text{ cm}^2$, 1200 lines/mm grating. The width of the proton beam, which the optical probe beam propagates through, is on the order of one hundred micrometers, and the transmission is assumed to be constant over this distance. Different wavelengths passed through the interaction area at

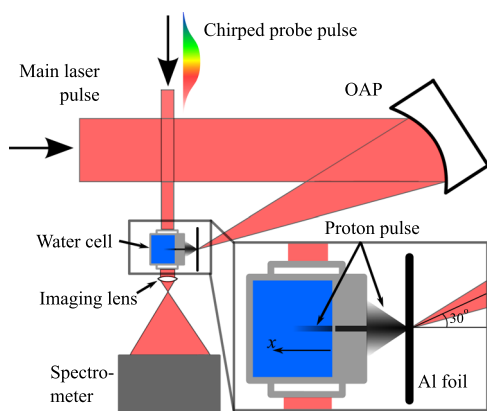


FIG. 1. Schematic sketch of the experimental setup. A high energy infrared laser pulse, compressed in the time domain to 600 fs (full width at half maximum), is focused by an off-axis parabolic mirror (OAP) onto a 12 μm thick Al target foil, at 30° angle of incidence. Protons are accelerated through the TNSA process. The ~ 0.1 mm central part of the proton pulse is allowed to enter through a 0.2 mm thick window of Teflon into a cell filled with water. The non-central protons are stopped in a several millimetre thick cover of Teflon. Perpendicular to the propagation direction of the protons in the water, along the x -axis, a chirped laser pulse is entering the water cell through a thin optical window. This pulse propagates through the volume where the accelerated protons interact with the water. Since the pulse is chirped, different frequencies will reach this volume at different times, thus giving a time resolution. The interaction volume is imaged and magnified by a lens onto the entrance slit of a Czerny-Turner imaging spectrometer. The main laser pulse and the probe pulse come from the same laser source and are therefore fully synchronised in time from shot to shot.

different times since the probe pulse was chirped. The wavelength bandwidth of the probe pulse is only a few nm, and its optical spectrum is fully enclosed in the much broader spectral region of absorption of the solvated electrons. The spectrometer then separated the frequencies spatially, thus creating a time-resolved image of the interaction. The images were captured at the exit of the spectrometer by a 16-bit CCD camera with 2048² pixels on a $27.6 \times 27.6 \text{ mm}^2$ sensor, with one axis corresponding to the time domain and the other axis the propagation depth of the laser-accelerated protons into the water. Unlike some other similar techniques that probe radiolytic processes with a chirped optical probe,^{17,18} our scheme includes an intrinsic temporal synchronization between the chirped pulse and a proton pulse. To determine the time resolution of the system, the spectrum of the chirped probe pulse was cut at a certain wavelength, thereby creating a step function in the wavelength domain. The 10%–90% rise time of the response in the spectrometer (giving the limit for the lowest resolvable time structures) was then corresponding to 12 ps \pm 1.5 ps for a probe pulse duration of 1 ns. In a similar way, the spatial resolution was determined, by imaging a sharp edge placed in the position of the water cell to be $2.5 \mu\text{m} \pm 0.5 \mu\text{m}$. Our experimental scheme thus allows for imaging both the temporal and spatial evolution, along the proton propagation axis, of the optical absorbance as the proton pulse propagates through the water cell.

For each set of data attained during the experiment, two recordings of the probe pulse were made: one with the proton pulse in the water and the other without. By comparing the two images, the change in transmission induced by the interaction between the high energy protons and the water could be deduced (Fig. 2). In the analysis process, the probe pulse intensities are set to be equal before the arrival of the proton pulse in the water cell to account for small fluctuations in the probe pulse intensity between the two recordings.

The absorbance, A , of a material is given by the relation

$$A = -\log_{10}(T) = \epsilon c l, \quad (1)$$

where T is the transmittance, ϵ denotes the molar attenuation coefficient, c is the absorbant concentration and l is the path length. In our case, the absorbing species, at the probe pulse wavelength, is solvated electrons. The concentration of solvated electrons mainly depends on the amount of energy deposited into the water by the laser accelerated protons but also somewhat on the linear energy transfer, i.e., the yield of solvated electrons is halved for protons close to the end of their tracks.⁶ The energy spectrum of the proton beam in these experiments is exponentially decaying, meaning that the highest amount of deposited energy per volume unit (highest dose) is found where the protons enter the water cell. This means that c and by extension A are in fact functions of the position along the proton propagation axis in the water cell, x .

The rising edge of the absorbance front, in other words the falling edge of transmission, was thoroughly investigated to confirm that the expansion velocity of the absorbance corresponds to the expected proton velocities. There is a variation in arrival time at the water cell with approximately 80 ps from the highest energy protons (~ 10 MeV corresponding to

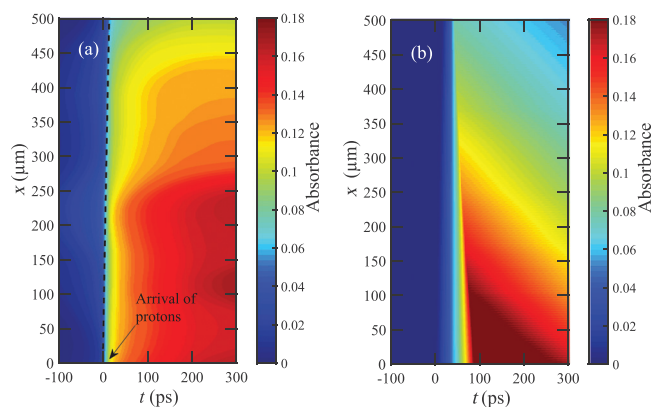


FIG. 2. (a) Experimental recording of induced absorbance in water to a chirped optical probe ($\lambda = 1053$ nm) when exposed to a pulse of high energy protons (arriving at $t = 0$ ps). The protons in the pulse have an exponentially decaying energy spectrum reaching up to approximately 10 MeV. The horizontal axis, t , represents the temporal evolution of the absorbance and the vertical axis, x , the spatial evolution along the axis of propagation of the energetic protons. The color scale indicates the level of absorbance, where 0 corresponds to full transmission. The dashed, black line shows how a proton with an initial energy of 10 MeV would propagate through the water. Small fluctuations in the spectrum of the probe pulse give high frequency noise in the analysis process. This has been removed from the image with an edge-preserving moving median-filter for illustrative purposes, but all analysis has been made with the unfiltered data. The image has been truncated from the full ns-duration of the probe pulse for clarity. (b) Model of absorbance in (a) based on the extrapolation of expected yields^{6,8} (approximately 4 solvated electrons per 100 eV of deposited energy, decreasing with increasing linear energy transfer) and decay kinetics^{19,20} of solvated electrons from studies with lower dose rates and measured at similar or longer times. The initial, exponentially decaying, proton energy spectrum is assumed to be similar to what was recorded during the experiment with a maximum proton energy of ~ 10 MeV. The point in time when the absorbance reaches its highest level for each value of x is determined by the decreasing proton pulse duration at increasing propagation depths.

$\sim 43 \mu\text{m/ps}$) to the lowest energy protons (~ 5 MeV corresponding to $\sim 32 \mu\text{m/ps}$) that can penetrate through the $200 \mu\text{m}$ entrance window of Teflon. When the proton pulse arrives inside the water cell, it is found that the absorbance first rises rapidly over the duration of the proton pulse, as expected from the swift processes that solvates electrons.²¹ The absorbance varies over the different propagation depths as expected from the discussion above, with the highest absorbance found where the highest dose is deposited.

A model has been developed to simulate how the absorbance changes as a function of depth in the water cell. The simulations are based on tabulated data from SRIM (The Stopping and Range of Ions in Matter)²² and the conversion efficiency from deposited energy into solvated electrons is taken from LaVerne *et al.*⁶ The molar attenuation coefficient, specific to our probe frequencies, is given in Assel *et al.*²³ The bandwidth of the chirped probe pulse is only a few nanometers and therefore the coefficient is, as a good approximation, constant throughout the whole pulse. Furthermore, the small decay of solvated electrons at room temperature on the picosecond timescale, as presented by De Waele *et al.* and El Omar *et al.*,^{19,20} is included in the model. The protons are also assumed to have an initial energy distribution, before propagating through the Teflon window, similar to what was measured during the experiment, i.e., an exponentially decaying proton energy spectrum from $\sim 10^{11}$ protons/MeV/sr at 1 MeV to $\sim 10^9$ protons/MeV/sr at 10 MeV. The results

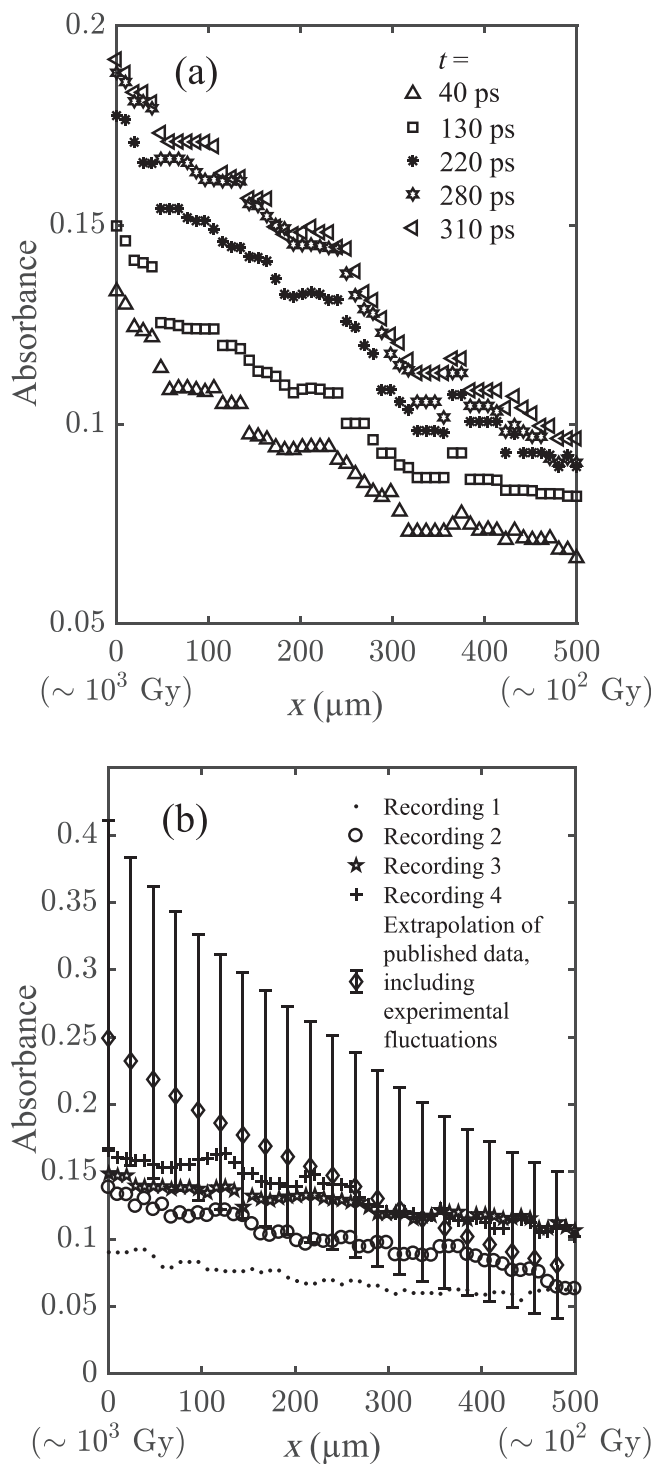


FIG. 3. (a) Absorbance of water as a function of the penetration depth, x , when exposed to a proton pulse at $t = 0$ ps, at five different times. The proton pulse duration is below 80 ps for all values of x , but a clear increase in absorption is measured up to at least 200 ps. This trend is clearer for low values of x , i.e., higher doses. (b) Absorbance of water at $t = 100$ ps as a function of the penetration depth, x , for four different recordings, when exposed to a proton pulse at $t = 0$ ps. The diamonds mark expected absorbance according to the model displayed in Fig. 2(b), based on the extrapolation of the yields of solvated electrons from publications^{6,8} with lower dose rates. The error bars take into account the shot-to-shot fluctuations in the proton energy spectra, in terms of both maximum energy and the number of protons for each energy. The shot to shot variations cannot fully explain the discrepancy between the model and the measured data for the highest doses, indicating that on the picosecond time scale, for sufficiently high dose rates, other physical processes take place compared to lower dose rates and longer time scales.

of these simulations are shown in Fig. 2 together with corresponding experimental recordings.

A comparison between the model and the experimental findings (see Fig. 3) shows reasonable agreement for the level of absorbance for the expected doses, especially for the lower doses. In Fig. 3(b), the error bars in the model represent likely shot-to-shot fluctuations in the proton energy spectra, both considering variations in maximum energy (20%) and the number of protons for each energy interval (50%). For lower doses, all the recordings fall within the error bars. For the highest doses, i.e., where the protons enter the water cell, there is however a clear discrepancy. One possible explanation for this is that the model does not take into account any intertrack effects. A high energy proton passing through water will deposit energy along its track in the so called spurs of radiolytic species. In this experiment, the track radius is in the order of a few nm²⁴ at the times considered. The area density, where the protons enter the water cell, is approximately 5000 protons/ μm^2 , indicating that the track structures of 5 nm radius would fill nearly half the water volume. Kreipl *et al.*²⁵ show through simulations a decrease in the yield of solvated electrons in the tracks of 20 MeV protons propagating through water, when the protons are in close proximity of one another. This effect increases with the temporal and spatial density of the proton tracks. Since the linear energy transfer for the protons of lower energy increases, giving an even higher density of radiolytic species in their tracks, it seems reasonable to assume that intertrack reactions would be at least as important for the protons of 10 MeV or below. This may explain why the difference in absorbance between the lowest and the highest dose is slightly smaller in the experiment than that predicted by the model, suggesting that for sufficiently high dose rates the yield of solvated electrons is decreased. Given the linear relation between absorbance and concentration of solvated electrons (see Eq. (1)), the comparison in Fig. 3(b) between our experimental data and the model, extrapolating data from the previous publications, indicates that the correction in the yields of solvated electrons for high dose rates (kGy/80 ps) could be as large as a factor of two.

The proton pulse duration is approximately 80 ps when entering the water, but also at later times the absorbance continues to slowly increase (see Fig. 3(a)), in contrast to what would be expected from the swift solvation process of electrons²¹ and the decay kinetics involved.^{19,20,26} Towards 300 ps, this increase levels out. To avoid shot-to-shot fluctuations affecting the results in Fig. 3(a), the data from each of the different times are taken from one single recording. This further confirms that an extrapolation of yields of solvated electrons from longer times and lower dose rates does not fully describe the scenario.

In conclusion, we have presented experimental data of how picosecond bunches of high energy protons interact with water. This enables further understanding of how ultra-high dose rates affect the energy deposition into water. Our

experimental scheme provides a unique tool to study high energy proton/water interactions and gather information that was previously primarily accessible through simulations.

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